A Multiscale Approach for the Understanding of Water Film Formation

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Introduction

- Reductive immobilization of toxic and radioactive metals by gaseous hydrogen sulfide is a promising technology for *in-situ* remediation of soils and groundwater (Fig. 1 & 2).
- Rate of chromium(VI) reduction by gaseous hydrogen sulfide in the vadose zone soil is controlled by gas phase humidity and soil particle size (Fig. 3);
- It is believed that water film formation on solid surfaces is needed for effective contaminant reduction and immobilization (Fig. 4).
- Molecular Dynamics (MD) Simulation is used to understand the mechanism of water film formation.

Fig. 1. Hanford Site, WA. – A chromium(VI) contamination sit from nuclear reactors nearby Columbia River.

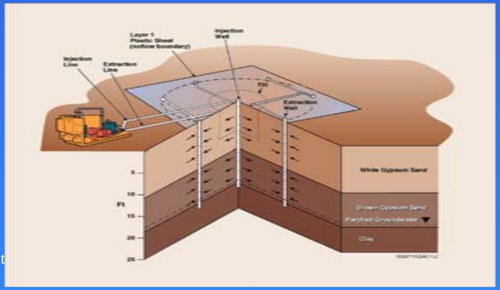

Fig. 2. *In-situ* gaseous reduction.

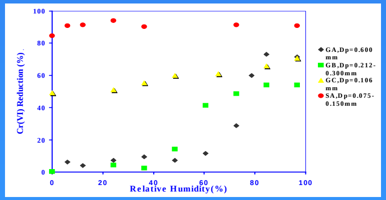

Fig. 3. Effects of humidity and particle size on chromium reduction.

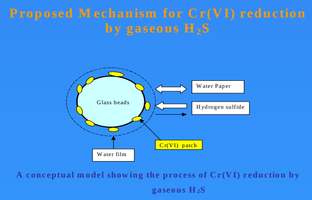

Fig. 4. A conceptual model for the formation of a layer of water film as the effective reaction zone.

Molecular Dynamics Simulation of Water Film Formation on Clay Surface

A classical molecular dynamic (MD) program LAMMPS was used to simulate water film formation on clay surfaces. The model TIP3P is used for water in the simulation. Interaction of water molecules with the substrate atoms is described by a potential model according to Rovere et al. (1998). Silicon atoms interact only via Coulombic forces with the charged water sites. The cross LJ parameters are calculated with the usual Lorentz-Berthelot rules. During the simulation the substrate atoms do not move. Ewald summation is performed to compute long-range Coulombic interactions. LJ and Coulombic interactions are computed with an additional switching function S(r) that ramps the energy and force smoothly to zero between 8 Å and 10 Å.

The simulation starts with the oxygen atoms of the water molecules placed in a FCC lattice. The size of the lattice is such that it is contained in the space above the clay surface. The initial velocities are chosen randomly with a distribution consistent with the required temperature. The water crystal is melted at 373 K and then is equilibrated at 373 K for 960 ps by using velocity rescaling technique. A time step size of 2 fs is applied throughout the simulation.

The MD simulation clearly captures the transition from water molecule clusters to water films (Fig. 5-6).

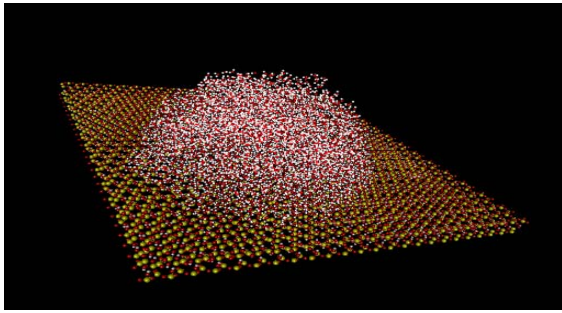

Fig. 5. A snapshot of MD simulation of 3600 water molecules on a patch of clay with a surface area of 127Å×128Å at t=1920×10^{-12}s.

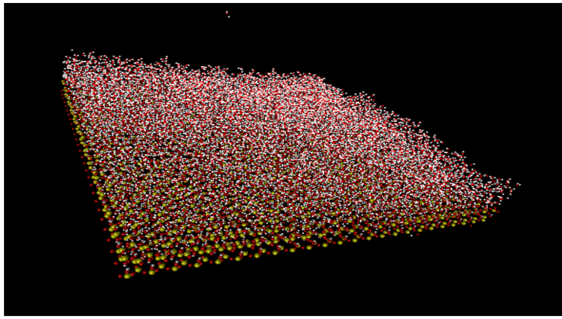

Fig. 6. A snapshot of MD simulation of 7200 water molecules on a patch of clay with a surface area of 127Å×128Å at t=1920×10^{-12}s.

Acknowledgements

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